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Dimethyl disulfide (DMDS) and dimethyl sulfide (DMS) emissions from biomass burning in Australia

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[1] We identify dimethyl disulfide (DMDS) as the major reduced sulfur-containing gas emitted from bushfires in Australia's Northern Territory. Like dimethyl sulfide (DMS), DMDS is oxidized in the atmosphere to sulfur dioxide (SO_2) and methane sulfonic acid (MSA), which are intermediates in the formation of sulfuric acid (H_2SO_4). The mixing ratios of DMDS and DMS were the highest we have ever detected, with maximum values of 113 and 35 ppbv, respectively, whereas background values were below the detection limit (10 pptv). Molar emission ratios relative to carbon monoxide (CO) were $[1.6 \pm 0.1] \times 10^{-5}$ and $[6.2 \pm 0.3] \times 10^{-6}$, for DMDS and DMS respectively, while molar emission ratios relative to carbon dioxide (CO_2) were $[4.7 \pm 0.4] \times 10^{-6}$ and $[1.4 \pm 0.4] \times 10^{-7}$, respectively. Assuming these observations are representative of biomass burning, we estimate that biomass burning could yield up to 175 Gg/yr of DMDS (119 Gg S/yr) and 13 Gg/yr of DMS. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0399 Atmospheric Composition and Structure: General or miscellaneous; *KEYWORDS:* Dimethyl disulfide, dimethyl sulfide, biomass burning, emission, Australia, BIBLE-B. **Citation:** Meinardi, S., I. J. Simpson, N. J. Blake, D. R. Blake, and F. S. Rowland, Dimethyl disulfide (DMDS) and dimethyl sulfide (DMS) emissions from biomass burning in Australia, *Geophys. Res. Lett.*, *30*(9), 1454, doi:10.1029/2003GL016967, 2003.

1. Introduction

[2] The emissions of trace gases from biomass burning have been studied extensively during the past two decades because of their potential impact on regional atmospheric chemistry and global climate change [*Crutzen et al.*, 1979; *Cofer et al.*, 1988; *Crutzen and Andreae*, 1990; *Bonsang et al.*, 1995; *Hao et al.*, 1996; *Koppmann et al.*, 1997]. Among the sulfur-containing gases, emphasis has previously been placed on the biomass burning yields of sulfur dioxide (SO_2) and carbonyl sulfide (OCS) [*Nguyen et al.*, 1995]. Very little is reported about dimethyl sulfide (DMS) emission from biomass burning [*Friedli et al.*, 2001] while no dimethyl disulfide (DMDS) emission estimates from biomass burning are currently available.

[3] OCS is the only reduced sulfur compound with an atmospheric lifetime long enough for a substantial fraction to be transported intact to the stratosphere (the lifetime of OCS is 2.5–4 years; *Johnson*, 1981). DMS has a much shorter lifetime ($\tau \sim 1$ day; *Lenschow et al.*, 1999) and DMDS has an even shorter atmospheric lifetime ($\tau \sim 0.3$–3 hours; *Hearn et al.*, 1990). The main removal process for DMDS is reaction with hydroxyl radicals (OH), with a reaction rate constant of $[1.98 \pm 0.18] \times 10^{-10}$ cm^3 molec^{-1} s^{-1} [*Wine et al.*, 1981]. DMDS also has minor removal by photolysis, 2.5×10^{-3} min^{-1} [*Tyndall and Ravishankara*, 1991] and by reaction with nitrate radicals (NO_3), $[7.0 \pm 2.0] \times 10^{-13}$ cm^3 molec^{-1} s^{-1}. However, removal by NO_3 may be significant at night due to the high nitrogen oxide (NO_x) levels produced in the fire, resulting in a shorter lifetime of the sulfur species under consideration. Because of its very rapid reaction with OH, DMDS has not been previously reported as a significant product from biomass burning. DMDS and DMS are quickly oxidized during daylight hours to compounds such as sulfur dioxide (SO_2) and methane sulfonic acid (MSA), which can play important roles in tropospheric chemistry, especially with regard to the formation of sulfuric acid (H_2SO_4) and subsequent formation of cloud condensation nuclei (CCN) [*Andreae and Crutzen*, 1997]. Here we investigate the emission of DMDS and DMS from bushfires in Northern Australia, with emphasis on their emission ratios.

2. Experimental

[4] For this study, 44 whole air samples were collected at ground level in Northern Australia during the late dry season as part of the BIomass Burning and Lightning Experiment-B (BIBLE-B). Three different fire episodes, two near Katherine (13°74′S, 131°73′E and 13°87′S, 131°84′E), and one further northeast in Arnhem Land (13°18′S, 133°58′E) were sampled between September 4–9, 1999. Each sample was collected in a conditioned, evacuated two-liter electropolished stainless steel canister equipped with a stainless steel bellows valve. Ground sampling was carried out very close to each fire (less than 3 meters away) during both the flaming and smoldering fire stages.

[5] The whole air samples were returned to our laboratory at the University of California, Irvine and analyzed for methane (CH_4), carbon monoxide (CO), carbon dioxide (CO_2), nonmethane hydrocarbons (NMHCs), halocarbons, sulfur gases, and alkyl nitrates. Methane and CO were analyzed using flame ionization detection (FID); CO_2 was analyzed using a thermal conductivity detector (TCD); nonmethane hydrocarbons were analyzed using FID and mass spectrometry (MS); and halocarbons and alkyl nitrates used electron capture detection (ECD) and MS.

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The analytical system used to quantify all gases except CO, CO_2 and CH_4 preconcentrated the sample on a stainless steel loop filled with glass beads and immersed in liquid nitrogen, with subsequent vaporization of the sample (see *Colman et al.* [2001] for complete analytical details). The sample was then flushed to a splitter that partitioned it to five different streams, with each stream sent to one of five column-detector combinations (DB1/FID, DB5/ECD, RESTEK1701/ECD, PLOT/FID and DB5/MSD). In order to determine the mixing ratios of the sulfur compounds in the biomass burning samples, the mass spectrometer was placed in the single ion monitoring (SIM) mode, choosing the most abundant ion of each compound without interference. Due to the complexity of the fire mixture, the analyses were performed using a 62-minute oven ramp in order to achieve a good resolution between peaks. Moreover, a peak purity analyses was performed on every single peak in order to ensure that interferences were avoided. The ions selected for the sulfur compounds were the main parent ions: OCS ion 60 m/z, DMS 62 m/z, and DMDS 94 m/z. The calibration stage was carried out after dilution of a Scott Marrin standard containing 0.943 ± 0.047 ppmv OCS; 1.017 ± 0.051 ppmv DMS; and 0.948 ± 0.047 ppmv DMDS. The precision, accuracy and limit of detection (LOD) for the sulfur compounds were 2%, 10%, and 10 pptv, respectively. For all NMHCs the precision, accuracy and LOD were 1%, 5% and 5 pptv, respectively.

3. Results and Discussion

[6] Enhanced DMDS and DMS mixing ratios were detected in the biomass burning samples collected during three fire episodes in Northern Australia (Table 1). Biomass burning in the Australian savanna is estimated to contribute 12–16% of the carbon released from tropical savanna fires [*Hurst et al.*, 1994]. Emission ratios for DMDS, DMS and OCS were calculated relative to CO and CO_2 in order to correlate their emission with the smoldering and the flaming stage of the fires, respectively. CO is mainly emitted during the smoldering stage when the temperature is usually lower than 850 K and the oxygen concentration can be as low as 5% [*Crutzen and Goldammer*, 1993]. On the contrary, CO_2 is mainly emitted during the flaming stage when the temperature is higher (>950 K) and the oxygen concentration is higher than 15%. Samples can be classified in terms of combustion efficiency [*Ward and Hardy*, 1991], which is defined as the percent carbon released in the form of CO_2. In previous work, samples collected during African savanna burning were classified in terms of the independent variable $\Delta CO/\Delta CO_2$ [*Bonsang et al.*, 1995]. To determine $\Delta CO/\Delta CO_2$ for the Northern Australian fires, average CO and CO_2 values of 6 background samples (103 ± 18 ppbv for CO and 370 ± 3 ppmv for CO_2) were subtracted from the CO and CO_2 mixing ratios measured for each fire sample. A $\Delta CO/\Delta CO_2$ ratio <0.1 ppmv/ppmv is typically observed for a high combustion efficiency (flaming) fire and is associated with high values of ethyne (C_2H_2) and ethene (C_2H_4). By contrast, a ratio >0.1 ppmv/ppmv indicates the smoldering stage of a fire [*Bonsang et al.*, 1991] and is associated with high alkane emissions. Applying the above criteria, 15 samples collected in the Northern Australian savanna fire plumes represented the smoldering stage of the fires and 20 samples represented the flaming stage of the fires.

Table 1. Minimum, Average and Observed Mixing Ratios of Selected Gases for Two Air Samples Collected During the Smoldering Phase of Fires in Katherine and Arnhem Land

	Mixing Ratio (ppbv)			
Compound	Katherine	Arnhem Land	Minimum	Average
CO_2	$2{,}570 \times 10^3$	$31{,}000 \times 10^3$	410×10^3	$3{,}820 \times 10^3$
CO	300×10^3	$6{,}080 \times 10^3$	198	698×10^3
Ethyne	753	148×10^3	0.431	8,592
Ethene	3,100	217×10^3	0.512	16.4×10^3
Ethane	1,100	26×10^3	0.867	2,887
CH_3Cl	577	1816	0.697	375
OCS	34	328	0.546	52
DMS	5.9	35	0.014	3.2
DMDS	22	113	0.254	15
CS_2	0.57	16	0.005	1.9

CH_3Cl - methyl chloride; CS_2 - carbon disulfide.

[7] The median DMS and DMDS mixing ratios measured in the fire plumes were 658 and 5,558 pptv respectively, while the mixing ratios of DMDS and DMS in the 6 background samples were below detection (<10 pptv). The highest mixing ratios of DMDS and DMS were measured in an Arnhem Land fire sample containing 113 ppbv of DMDS and 35 ppbv of DMS, with CO and CO_2 concentrations of 6,080 ppmv and 31,000 ppmv, respectively (Table 1). This DMS mixing ratio is the highest ever measured by our group, and it is 100 times higher than maximum mixing ratios (370 pptv) recorded over productive oceanic regions during the PEM-Tropics B experiment [*Simpson et al.*, 2001]. More importantly, this paper reports the first quantitative measurements of DMDS emission ratios from biomass burning.

[8] The mixing ratios of many other gases were also relatively high in the fire samples (Table 1). Maximum values of CO and other hydrocarbons were much lower for the Katherine samples than for the Arnhem Land samples. In order to determine if the Northern Australia fires emission could be considered representative of biomass burning in general, plots of all the BIBLE-B NMHC fire data versus CO were made [*Shirai et al.*, 2003], yielding good correlation coefficient values ($R^2 > 0.9$) and similar emission ratios relative to CO for most NMHCs compared to previously measured values in Brazil and Africa [*Blake et al.*, 1996]. However ethane is more than a factor of two lower than the corresponding values from Brazil and Africa. Similarly, propane and *n*-butane have smaller emission ratios than in the Brazil and Africa samples, suggesting a larger flaming influence in the Australian samples. In addition, the highly reactive species propene (C_3H_6) is elevated relative to CO compared to the African and Brazilian samples, most likely because of the much closer proximity of the Australian sampling to the fires.

[9] Plotting DMDS versus CO for the smoldering samples collected in Northern Australia yields an average emission ratio of $[1.6 \pm 0.1] \times 10^{-2}$ ppbv/ppmv. The DMS versus CO emission ratio has a lower slope of $[6.2 \pm 0.3] \times 10^{-3}$ ppbv/ppmv. Different patterns are recognizable for DMDS and DMS emissions during the flaming stage of the fires, calculated with respect to CO_2. DMDS correlated well with

Table 2. DMS and DMDS Emission Estimates From Savanna Fires and All Types of Biomass Burning Worldwide

	Emission Ratio, $\times 10^{-6}$	[a]Savanna Reference Species Flux, Tg yr^{-1}	All Savanna Emissions Gg yr^{-1}	[b]Worldwide Reference Species Flux, Tg yr^{-1}	All Biomass Burning Emissions, Gg yr^{-1}
$\Delta DMS/\Delta CO$	6.2	240	3.3	748	10
$\Delta DMS/\Delta CO_2$	0.14	6,090	1.2	13,400	2.6
$\Delta DMDS/\Delta CO$	16	240	14	748	41
$\Delta DMDS/\Delta CO_2$	4.7	6,090	61	13,400	134

[a]Emission rates of CO and CO_2 from savanna burning proposed by *Andreae et al.* [1996].
[b]Worldwide global pyrogenic emission of CO [*Holloway et al.*, 2000] and CO_2 [*Andreae and Merlet*, 2001].

CO_2 and had a slope of $[4.7 \pm 0.4] \times 10^{-3}$ ppbv/ppmv, while DMS versus CO_2 had a slope of $[1.4 \pm 0.4] \times 10^{-4}$ ppbv/ppmv (Table 2). The generally good correlation of DMDS with both CO ($R^2 = 0.91$) and CO_2 ($R^2 = 0.91$) suggests that it is emitted during both the smoldering and flaming stages of the fire. On the contrary, the good correlation of DMS with CO ($R^2 = 0.94$) and a poorer correlation with CO_2 ($R^2 = 0.47$) indicate that DMS is mainly emitted during the smoldering stage of the fire. These results point to different combustion chemistries for the formation of DMDS and DMS.

[10] In order to estimate the magnitude of total sulfur gas emissions from biomass burning, a comparison with OCS emissions was carried out. During the smoldering stage, OCS versus CO produced a slope of $[5.4 \pm 0.3] \times 10^{-2}$ ppbv/ppmv, while no significant correlation with CO_2 was observed. This value is within a factor of two of the emission ratios reported for biomass burning in Africa and South East Asia (8.5×10^{-2} ppbv/ppmv; *Nguyen et al.*, 1995) and in the western United States (9.0×10^{-2} ppbv/ppmv; *Friedli et al.*, 2001).

[11] We employed our biomass burning emission ratios of DMDS and DMS to estimate tropical emissions from savanna fires and also for worldwide biomass burning. These calculations assume that the Australian savanna fire samples are representative of biomass burning in general. This assumption is somewhat supported by the similar NMHC emission ratios for these Australian fires compared to those calculated for African and Brazilian fires [*Blake et al.*, 1996], although emission ratios also depend on fuel composition. We are not aware of any elemental composition analyses of fuel sulfur from the Australian savanna, which is mainly composed of *Triodia species* (common name *Spinifex*); *Sandstone Heath*, a highly flammable community that contains many fire sensitive species; *Sorghum species*, more often known as long grass as it grows usually to about three meters tall; and two different kinds of *Eucalyptus* (*Eucalyptus tectifica* and *Eucalyptus latifolia*) with a *Sorghum* grassland understorey. In addition, because significant uncertainties exist for biomass burning emission estimates of CO and CO_2 (used in these calculations), the uncertainties in the emission estimates of DMDS and DMS are probably at least a factor of 2.

[12] *Andreae et al.* [1996] suggest that savanna fires produce a total of 6,090 Tg CO_2 yr^{-1} with a mean $\Delta CO/\Delta CO_2$ ratio of 0.063 (corresponding to a CO flux of 240 Tg yr^{-1}; *Ward et al.*, 1992; *Lacaux et al.*, 1995). Scaling up our emission ratios employing these values gives estimates for DMDS and DMS emissions for savanna burning of 75 Gg yr^{-1} and 4.5 Gg yr^{-1}, respectively (Table 2). These upper limit estimates are obtained by adding the separate emission ratios versus CO and CO_2, representing emissions during the smoldering and flaming stages of the fires. Similarly, global pyrogenic emissions of 13,400 Tg CO_2 yr^{-1} [*Andreae and Merlet*, 2001] and 748 Tg CO yr^{-1} [*Holloway et al.*, 2000] were used to obtain the global emission estimates from all biomass burning (175 Gg yr^{-1} for DMDS and 12.6 Gg yr^{-1} for DMS; Table 2). The DMS emission estimate from biomass burning is small compared to a global marine emission estimate of 24.5 Tg yr^{-1} [*Watts*, 2000].

[13] Our calculated emissions for OCS, DMS, and DMDS correspond to a global sulfur emission of 0.171 Tg S yr^{-1} from biomass burning (Table 3). Because each DMDS molecule contains 2 sulfur atoms, sulfur emission from DMDS, calculated with respect to CO_2 for the flaming stage of the fire, is approximately 2.5 times larger than that from OCS during the smoldering stage (no OCS emission during the flaming stage was detected).

[14] In the atmosphere, both DMS and DMDS are rapidly oxidized to SO_2 and MSA, which will subsequently lead to H_2SO_4 or undergo dry deposition. Biomass burning is a significant direct source of SO_2 to the atmosphere. Annual global biomass burning SO_2 emissions are currently estimated to be about 3.5 Tg (or 1.8 Tg S) [*Andreae and Merlet*, 2001]. Thus, DMDS, DMS and OCS emitted during biomass burning could account for a further 10% contribution to atmospheric sulfur released annually from biomass burning. About half of these emissions are in the form of DMDS.

[15] Airborne measurements of DMS and DMDS during BIBLE-B showed elevated DMS mixing ratios during two flights that encountered biomass burning plumes [*Shirai et al.*, 2003], suggesting that DMS (which is longer-lived than DMDS) can be exported into the free troposphere, thereby escaping the local burning environment. However, the

Table 3. Global Emission Estimates of Sulfur Released to the Atmosphere in the Form of DMDS, DMS and OCS During Biomass Burning

	Emission estimate Tg S yr^{-1}
S from OCS: This work	0.046
Nguyen et al. [1995]	0.03–0.11
Crutzen et al. [1979]	0.05–0.27
Bingemer et al. [1992]	0.05–0.35
Yokelson et al. [1997]	0.033
Friedli et al. [2001]	0.075–0.10
S from DMDS: This work	0.119
S from DMS: This work	0.006
Friedli et al. [2001]	0.006–0.019
Total S from DMDS + DMS: This work	0.125
Total S from DMDS + DMS + OCS: This work	0.171

The estimates are based on global biomass burning emissions of 748 Tg yr^{-1} for CO and 13,400 Tg yr^{-1} for CO_2.

mixing ratios encountered during the airborne portion of BIBLE-B were much smaller than those measured on the ground during this study, suggesting that the impact of DMDS and DMS emissions from biomass burning is largely confined to the boundary layer. Further, the rapid oxidation of DMDS and DMS to form SO_2 is probably localized by the high aerosol surface area loading in the biomass burning air masses. Thus, DMDS and DMS oxidation may provide an important pathway for returning organic sulfur to the biosphere as nutrient sulfate.

4. Conclusions

[16] Savanna biomass burning is a major localized source of DMDS and DMS. The average mixing ratios of these two sulfur compounds in fires plumes in Northern Australia are by far the highest ever reported in the troposphere. These data represent the first emission estimate of DMDS reported from biomass burning. DMDS is emitted during both the flaming and smoldering stage of the fire, in contrast to DMS and OCS which are mainly emitted during the smoldering stage. Although DMS emission from biomass burning has a small impact on the total budget of DMS worldwide, together these three reduced sulfur species are estimated to account for a further 10% of the atmospheric sulfur emitted by biomass burning. The emissions of DMDS and DMS appear to be largely confined to the boundary layer, and their rapid oxidation to SO_2 may be an important path for returning organic sulfur to the biosphere. The identification of DMDS and DMS at significant concentrations provides a useful addition to our understanding about processing of sulfur in biomass burning.

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